A bond contact model for methane hydrate bearing sediments with inter-particle cementation

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Abstract

While methane hydrate (MH) can be present in various forms sands deep seabeds or permafrost regions, this paper deals with methane hydrate bearing sediments (MHBS) where the MH has formed bonds between sand grains. A bond model based on experimentally-validated contact laws for cemented granules is introduced to describe the mechanical behavior of the MH bonds. The model parameters were derived from measured values of temperature, water pressure and MH density. Bond width and thickness adopted for each bond of the MHBS were selected based on the degree of MH saturation. The model was implemented into a 2D-DEM code. A series of numerical biaxial standard compression tests were carried out for various degrees of MH saturation. Comparison with available experimental data shows that the model can effectively capture the essential features of the mechanical behavior of MHBS for a wide range of levels of hydrate saturation. In addition, the analyses here presented shed light on the relationship between level of cementation and de-bonding mechanisms taking place at the microscopic level and the observed macro-mechanical behavior of MHBS. Also the analyses shed light on the relationship between spatial distribution of bond breakages and contact force chains with the observed strength, dilatancy and deformability of the samples.

Keywords: methane hydrate; cementation; bond contact model; discrete element method; biaxial compression tests

1 Introduction

Methane hydrate (MH) is regarded as one of the most promising resources to alleviate current and future energy needs [1-4]. MHs usually form in deep seabeds and permafrost regions where MHs can remain stable under low temperatures and high pressures [5-8]. They can greatly enhance the strength of the host sediments [9]. However, changes in pressure and temperature conditions and human interventions (i.e., installation of offshore pipelines, cables and platforms) may cause MH dissociation with the consequent destabilization of methane hydrate bearing sediments (MHBS) and lead to large marine landslides. Unfortunately, the mechanisms originating these geo-hazards are still poorly understood due to a lack of knowledge of the fundamental mechanical properties of MHBS. This also makes it difficult to establish a universal constitutive model for MHBS [8], although some models were proposed in the literature showing capability in capturing several basic features of MHBS (e.g., [10, 11]), and were employed to solve boundary value problems using either the finite element method (FEM) or the finite difference method (FDM) (e.g., [12]).

The formation of MHs is affected by a few factors such as host deposit, gas percolation speed and path, and initial water saturation [13, 14], leading to a variety of micro structures in MHBS. As illustrated in Figure 1, Waite et al. [15] identified three main formation habits at the pore scale: (1) pore-filling, with MHs floating in the pore fluid without bridging any particles; (2) load-bearing, with hydrate particles taking part in the strong force chains of the granular assembly; and (3) cementation, with

MHs cementing sand grains (acting as bond bridges between grains). As suggested by Waite et al. [16] and confirmed by experimental data from different sources(e.g., [13, 17, 18]), the mechanical properties of MHBS strongly depend on the formation habits of the hydrates. For instance, hydrates acting as cementation agents at inter-particle contacts give rise to larger values of strength and stiffness for the MHBS than pore-filling hydrates. This implies that hydrate morphology has to be taken into account in any realistic constitutive models of MHBS. Only an investigation at the pore scale can establish the link between pore habits and macroscopic properties of MHBS (e.g., [19]).

The distinct element method (DEM), originally proposed by Cundall and Strack [20] for dry granules, has significant potential in shedding light on the relation between pore habit of the hydrates and bulk properties of MHBS by modeling hydrate morphology at the grain scale. Recently, DEM has been employed to investigate some significant features of MHs, such as hydrate growth [21], hydrate dissociation [22], hydrate distribution of pore-filling patterns [8, 23, 24] or of cementation patterns [25, 26]. Brugada et al. [8] investigated the mechanical properties of MHBS with MH in the form of pore-filling habit via three-dimensional DEM simulations. Discrete spheres one order of magnitude smaller than the soil particles were randomly generated and distributed within the voids to replicate hydrate particles. In their simulations, soil grains and MH particles get into contact and exchange forces, however without exhibiting any cementation at contacts. In that work, the contact stiffnesses of the hydrate particles were determined on the basis of a parametric study.

Jung et al. [23, 24] characterized mechanical behaviors of MHBS in two cases: distributed hydrates and patchy saturation. In these works hydrate particles were randomly generated in space and attached to the sand grains so that their effect on the load bearing force chains could be investigated. However, few researches have been carried out on the cementation pattern of MHs due in part to a lack of properly validated bond contact models for MHBS accounting for essential factors such as hydrate saturation, temperature, and water pressure. Although some early attempts were made to model MHs in the cementation habit via DEM, the previous models were over-simplified without clarifying the exact correlation between hydrate saturation and bond strength [25], or neglecting the role played by the thickness of the MH bonds [26]. However, MHs forming bonds between grains of the host granular soil are of common occurrence since MHs are prone to cement unconsolidated sediments containing an abundant gas phase. For example, in the Blake Ridge off the southeast coast of the United States [27] and in the Cascadia margin [28], MHs have been recognized in formation habit as cementing. In this case, the formation of MHs causes the onset of hydrate bonds between nearby sand grains at the pore scale. Experimental data [14, 16, 29-32] show that this pore habit plays an important role in the macro-mechanical behavior of MHBS, and in particular substantially affects the bulk properties of MHBS far more than the pore-filling habit. Therefore there is a need to investigate the influence of MH bonds on the bulk properties of the host sediments. To achieve this goal via DEM simulations, a suitable bond contact model accounting for all the significant factors affecting the bond behavior is of critical

67 importance. Establishing a suitable bond model also paves the way to use DEM
68 analyses to investigate the effect of time dependent phenomena taking place within
69 inter-particle bonds on MHBS. In fact, chemical reactions may occur over time with
70 the effect of strengthening or weakening the bonds (aging effects). The DEM could be
71 employed to investigate these time-dependent phenomena.

72 It has been widely recognized that inter-particle bonds in some other cemented
73 materials, such as sandstone, mortars, grouted soils and volcanic ashes, affect many
74 aspects of soil behavior, e.g., enhancing strength and shear dilation [33, 34]; ruling
75 strain softening [35-41]; and influencing the formation of shear bands [38, 42]. The
76 inter-particle bonds of these cemented soils are mainly made of Portland cement,
77 gypsum and lime owning properties different from those of MHs. Little is known
78 about MH bonds which, however, largely affect the macro-mechanical behavior of
79 MHBS. Hence, it is authors' opinion that research on the macro-mechanical
80 properties of MHBS should be informed by its microstructure and the behavior
81 observed at the micro scale. Scanning electron microscopy (SEM) images and X-ray
82 CT images have been used to obtain morphological information of MHBS at the scale
83 of the grain. However, these techniques alone are insufficient to gain a comprehensive
84 understanding of the macro mechanical response of MHBS, particularly in relation to
85 the bonding effects of MHs.

86 This paper aims to establish a suitable bond contact model for sandy deposits
87 with MH bonds based on an experimentally-validated contact law achieved for
88 cemented granules. Following this introduction, a general bond contact model derived

89 from a series of micro experiments is introduced with particular emphasis on the bond
90 strength envelope. Section 3 extends the generic model to MHBS by relating the
91 model parameters to the surrounding temperature, water pressure, and MH density in
92 addition to the hydrate saturation. DEM implementation of this model is described in
93 Section 4 followed by relevant simulation results to be presented in Section 5 in
94 comparison to the available experimental data [18].
95

96 ## 2. A bond contact model for cemented granules

97 Figure 2 illustrates conceptually the 2D bond contact model adopted in the DEM
98 simulations presented in this paper. The bond between two disks of radii R_1 and R_2
99 has a finite width, B, and a finite thickness varying along the disk surface. The bond
100 thickness is hereafter characterized by its measure at the center of the bond, t_0 (see
101 Figures 2(a)). Figures 2(b) to (d) provide the mechanical responses of this model in
102 three directions (i.e., normal, tangential and rolling directions). As illustrated in the
103 figures, the force-displacement and the moment-rotation laws are featured by an
104 initially linear elastic response, brittle breakage and perfect plastic behavior. This
105 laws have been experimentally derived from bonds made of either epoxy resin [43] or
106 cement [44].

107 For an intact bond, the normal force F_n, the shear force F_s, and the moment M can
108 be computed as:

109
$$F_n = K_n(u_n - u_0), \tag{1a}$$

110
$$F_s = K_s u_s, \tag{1b}$$

$$M = K_r \theta, \tag{1c}$$

where u_n, u_s, and θ are the overlap, relative shear displacement, and relative rotation angle, respectively, whilst K_n, K_s, K_r are the normal, tangential and rolling bond contact stiffness, respectively. u_0 is the distance between two adjacent particles at the time of formation of the bond. In the case that two particles are in contact during the formation of the bond, the minimum bond thickness, t_0, is assumed to be 0 and u_0= 0. To account for the distance existing between particles at the time of formation of the MH bonds is important for a realistic modeling of MHBS since this has an influence on the observed mechanical behavior of the assembly of bonded particles.

The thresholds (or bond strengths) in the normal, shear and rolling directions are here denoted by R_{nb}, R_{sb} and R_{rb}, which define the upper bound of the elastic regime in each direction. The bond will break in a fragile fashion if any force or moment exceeds these thresholds. When two grains after their bond has broken come into contact again, the contact laws between un-bonded grains are as follows:

$$F_n = K_n' u_n, \tag{2a}$$

$$F_s = \min\left[K_s' u_s,\ \mu F_n\right], \tag{2b}$$

$$M = \min\left[K_r' \theta,\ \frac{F_n \cdot \delta \cdot \bar{R}}{6}\right], \tag{2c}$$

where min[·] is the operator taking the minimum value; μ is the inter-particle friction coefficient; $\bar{R} = 2R_1R_2/(R_1+R_2)$ is the equivalent radius of two particles in contact. K_n', K_s' and $K_r' = K_n'(\delta\bar{R})^2/12$ are the normal, tangential and rolling contact stiffnesses between soil particles in contact which are different from the stiffnesses introduced in Eqs. (1) which account for both bond and particle stiffnesses. δ is the

shape parameter of soil particles characterizing the rolling resistance of particles in contact, which is meant to account for the effect of non-spherical grain shapes. Details on the physical meaning of the inter-particle rolling resistance can be found in Jiang et al. [45].

In case of combined loads, the adopted strength criterion is represented as a three-dimensional surface in the F_n-F_s-M space. According to experimental results on epoxy resin and cement [43, 44], the slice of the surface in the F_s-M plane can be described in first approximation by an ellipse:

$$\frac{M^2}{R_{rb0}^{\ 2}} + \frac{F_s^{\ 2}}{R_{sb0}^{\ 2}} = 1, \tag{3}$$

where R_{sb0} is the bond shear strength in the absence of bending moments and R_{rb0} is the bond rolling strength in the absence of shear forces. The size of the ellipse depends on F_n. Hence both R_{sb0} and R_{rb0} depend on the magnitude of F_n:

$$R_{sb0} = f_s \cdot L_s \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^n, \tag{4a}$$

$$R_{rb0} = f_r \cdot L_r \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^m, \tag{4b}$$

where R_{tb}, R_{cb} are the bond tensile and compressive strength, respectively, which can be obtained from pure tension and compression tests on the cemented granules. L_s, L_r are the slope of the straight lines linking R_{tb} to the peak shear strength or rolling resistance on the projection plane (see Figure 3). Coefficients f_s, f_r, n and m are fitting parameters calibrated on the available experimental data. A comparison between the curves of Equation (4) and available experimental data, [43, 44], is shown in Figure 3. A good agreement between the curves and the experimental data is

154 apparent. Combining Equations (3) and (4) gives rise to a three dimensional strength
155 envelope shown in Figure 4 which has proved to replicate satisfactorily the available
156 experimental data for combined loading paths too.

157 The contact law for an intact bond requires knowledge of the bond strength and
158 stiffness parameters summarized in Table 1. For cement- or epoxy-bonded case, these
159 parameters can be directly obtained from microscopic mechanical tests [43, 44]. For
160 the MHBS case, however, it is very difficult to conduct micro-mechanical tests on
161 MH bonded granules because of the extreme experimental conditions required, e.g.,
162 very low temperature and very high pressure, which make it very difficult to obtain
163 reliable direct measurements of the parameters of the MH bonds. Jung and
164 Santamarina [46] performed experimental tests at the micro scale to measure adhesive
165 and tensile strengths of two MH-bonded flat smooth surfaces of mica and calcite;
166 however their measurements are not directly applicable to the bonds considered here
167 since the bond strength is likely to be significantly affected by the curvature of the
168 surfaces of grains near their contact point, grain surface roughness and impurities in
169 the MH. Thus, it is necessary to introduce some assumptions to determine indirectly
170 the model parameters subsequently illustrated here.
171

172 ## 3. Extension of the bond contact model to MHBS

173 Concerning MHBS located at a depth of h below sea level, Figure 5(a) provides a
174 sketch of two MH bonded granules at the temperature T, and the surrounding water
175 pressure σ_w. In this section, the model parameters (i.e. bond strength and bond contact

176 stiffness) relevant to MH bonds will be indirectly determined with respect to the
177 surrounding environment. Moreover, the geometric features (e.g, the width and
178 thickness) of the MH bonds will be studied considering the MH saturation and MH
179 formation characteristics in the host sands.

180 **3.1 Bond contact stiffness of MH**

181 As shown in Equation (1), the bond contact stiffness can be characterized by K_n, K_s,
182 K_r in the normal, shear and rolling direction, respectively. In general, the Young's
183 modulus for soil grains ranges from 50 to 70 GPa. They can be regarded as rigid
184 particles when the bonds (e.g., MH bonds) have relatively lower elastic modulus. As
185 shown in Figure 2, for the case of rigid particles with deformable elastic bond, the
186 normal deformation of the bond material, δ_n, can be expressed as:

187 $$\delta_n = \varepsilon t = \frac{\sigma t}{E} = \frac{F_n t}{BE}, \tag{5}$$

188 where ε is the normal strain; σ is the evenly distributed normal stress; E is the
189 Young's modulus of the bonding material and t is the average thickness of the
190 bonding material (see Figure 2). Thus the normal contact stiffness can be related to E
191 as follows:

192 $$K_n = \frac{F_n}{\delta_n} = \frac{BE}{t}. \tag{6}$$

193 According to experimental data obtained from tests on pure MHs (e.g., [47]), E
194 is strongly related to the temperature, T, the confining pressure, (i.e., pore pressure in
195 this case, σ_w), and MH density, ρ. The relationship can be written as:

196 $$E = E(\sigma_w, T, \rho) \quad . \tag{7}$$

Figures 6(a) and (b) present the collected stress-strain response curves obtained from triaxial compression tests on pure MH samples under different testing temperatures and confining pressures [47]. The information available on MH density is also presented for each curve. The Young's modulus of MH under different testing conditions, i.e., the tangential modulus at the point where the deviator stress is one half of the peak value, was obtained and listed in Table 2, which was used to produce Figure 7 in order to show the factors influencing the Young's modulus of MH. As shown in Figs. 7(a) and (b), the Young's modulus increases linearly with the MH density at a rate which is not significantly affected by confining pressure and temperature. Data in Figs. 7(a) and (b) can be regrouped for different MH densities (i.e., 0.7, 0.8 and 0.9 g/cm^3) resulting in Figs. 7(c) and (d) that illustrate the variation of the Young's modulus of MH with respect to confining pressure and temperature. Figures 7(c) and (d) indicate that the Young's modulus of MH linearly increases with confining pressure and linearly decreases with temperature at a rate which is significantly affected by the MH density. Hence we assume absence of coupling between the three factors: confining pressure, temperature and MH density. The Young's modulus of MH can be expressed as:

$$\frac{E}{p_{\mathrm{a}}} = a_0 + a_1\left(\frac{\sigma_w}{p_{\mathrm{a}}}\right) + a_2\left(\frac{T}{T_0}\right) + a_3\left(\frac{\rho}{\rho_w}\right), \tag{8}$$

where p_a is the standard atmospheric pressure (i.e., 1.01×10^5 Pa); T_0 is a reference temperature of 1 ˚C; ρ_w is the density of water at the temperature of 4˚C; a_0, a_1, a_2, and a_3 are constant coefficients achieved by fitting the data in Table 2. As a result, Equation (8) yields:

$$\frac{E}{p_a} = 3\left(\frac{\sigma_w}{p_a}\right) - 1.98\left(\frac{T}{T_0}\right) + 4950.50\left(\frac{\rho}{\rho_w}\right) - 1821.78 . \quad (9)$$

The values of Young modulus obtained by using Equation (9) are listed in Table 2 for sake of comparison with the available experimental data. A good agreement is apparent. The tangential and rolling bond contact stiffnesses can be assumed to be proportional to the normal contact stiffness, with the former one determined by experimental investigations [43, 44] and the latter one determined by the assumption of elastic bond, [36]:

$$K_s = \frac{2}{3} K_n \quad . \quad (10)$$

$$K_m = \frac{1}{12} K_n B^2 . \quad (11)$$

3.2 Bond strengths of MH

3.2.1 Bond tensile/compressive strength

When a tension or compression force is applied, the normal force is assumed to be evenly distributed over the whole width of the inter-particle MH. Hence, tensile and compressive strengths of MH bonds, i.e. R_{tb} and R_{cb} in Equations (4), can be computed from the tensile and compressive strengths of a pure MH specimen, $q_{\max,t}$ and $q_{\max,c}$ respectively, subject to a given confining pressure σ_w:

$$R_{tb} = B \times q_{\max,t} , \quad (12a)$$

$$R_{cb} = B \times q_{\max,c} , \quad (12b)$$

Figure 5(b) provides the shear strength envelope of pure MH having assumed the validity of the Mohr-Coulomb failure criterion. Given a prescribed pore water

pressure, σ_w, the minimum and maximum principal stresses are σ_t and σ_c, respectively. Depending on the value of water pressure, σ_t may also be tensile (i.e. negative according to the soil mechanics stress convention). Thus, $q_{\max,t}$ and $q_{\max,c}$ can be expressed as:

$$q_{\max,t} = (\sigma_w - \sigma_t), \tag{13a}$$

$$q_{\max,c} = (\sigma_c - \sigma_w), \tag{13b}$$

The maximum deviator stress $q_{\max,c}$ obtained from a compression triaxial test on a pure MH specimen (without any soil grains included) strongly depends on the temperature, T, confining pressure , σ_w, and MH density , ρ [47]. Hence it can be expressed as:

$$q_{\max,c} = q_{\max,c}(\sigma_w, T, \rho), \tag{14}$$

so that in the light of Equations (12), $R_{tb} = R_{tb}(\sigma_w, T, \rho)$ and $R_{cb} = R_{cb}(\sigma_w, T, \rho)$. Figures 8(a), (b) and (c) illustrate the variation of the maximum deviator stress against pore water pressure, temperature and MH density based on published experimental data [47]. In Figure 8 (b), the solid line and dash line represent the experimental results of MH with high and low purity, respectively. Results of MH at high purity were selected in our analysis. Since the tests on MH with high purity were only performed at T=-30 °C, the dash line is plotted assuming that it exhibits the same tendency as that of the solid line. As shown in these figures, the maximum deviator stress of MH linearly increases with the MH density and the confining pressure, and linearly decreases with the temperature. For sake of simplicity, we neglected the reciprocal influence of the three factors on the maximum deviator stress of MH. Thus,

Equation (14) can be expressed by:

$$\frac{q_{\max,c}}{p_a} = b_0 + b_1\left(\frac{\sigma_w}{p_a}\right) + b_2\left(\frac{T}{T_0}\right) + b_3\left(\frac{\rho}{\rho_w}\right), \quad (15)$$

where b_0, b_1, b_2, and b_3 are fitting parameters. Table 3 lists the available experimental data of MH with high purity [47] and the data attained from the dash line in Figure 8(b). Fitting the data in Table 3, Equation (15) yields:

$$\frac{q_{\max,c}}{p_a} = 0.81\left(\frac{\sigma_w}{p_a}\right) - 2.08\left(\frac{T}{T_0}\right) + 184.16\left(\frac{\rho}{\rho_w}\right) - 134.65. \quad (16)$$

The prediction based on Equation (16) is also given in Table 3 for sake of comparison with the available experimental data. A good agreement is apparent. Equation (16) is assumed to also hold for the extension triaxial test, leading to the following:

$$\frac{q_{\max,t}}{p_a} = 0.81\left(\frac{\sigma_t}{p_a}\right) - 2.08\left(\frac{T}{T_0}\right) + 184.16\left(\frac{\rho}{\rho_w}\right) - 134.65. \quad (17)$$

Combining Equation (13a) and Equation (16) to cancel out σ_t, Equation (17) can be re-written as:

$$\frac{q_{\max,t}}{p_a} = 0.45\left(\frac{\sigma_w}{p_a}\right) - 1.15\left(\frac{T}{T_0}\right) + 101.75\left(\frac{\rho}{\rho_w}\right) - 74.39. \quad (18)$$

3.2.2 Shape of the strength envelope

As demonstrated in Figure 3, the shape of the envelope is controlled by the cementation materials, resulting in different values of fitting parameters in Equation (4). Direct calibration of these parameters from laboratory tests for MH bonds in the same manner as cement or epoxy bonds [43, 44] is extremely difficult. So at present

282 the mechanical properties of MH-bonded sand grains can only be inferred by standard
283 macroscopic geotechnical tests and analogy to similar types of materials. In Figure 9,
284 the yielding curves of different materials in the normalized $(\sigma_1 - \sigma_3)$ and σ_3 plane
285 are presented. The yielding curve is 'right skewed' with a right tail for cement-based
286 material and 'left skewed' for epoxy resin. Unfortunately, the yielding curve of MH
287 cannot be inferred from the scanty experimental data available. However, MH is an
288 ice-like material composed of methane gas and water, i.e., natural gas is trapped
289 inside cage-like crystal structures made up of water molecules [7]. Its physical [48, 49]
290 and mechanical [50, 51] properties have been found to be similar to those of ice.
291 Therefore the yielding curve of ice has been plotted in Figure 9(b) for comparison.
292 The curve relative to ice is left skewed akin to cement-based materials. Hence, it can
293 be inferred that the tangential/rolling bond strength envelope of MH resembles that of
294 cement-based materials. Accordingly, the fitting parameters determined for the shape
295 of the strength envelope in case of cement bonds [44] can be used for MH bonds.
296 Thus, Equations (4) for MH bonds are here re-written as:

297 $$R_{sb0} = 1.38 \cdot 0.38 \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^{0.59}, \quad (19a)$$

298 $$R_{rb0} = 1.366 \cdot 0.741 \cdot (F_n + R_{tb}) \cdot \left(\ln \frac{R_{cb} + R_{tb}}{F_n + R_{tb}} \right)^{0.59}, \quad (19b)$$

299 where R_{tb} and R_{cb} can be computed from a combination of Equations (12), (16) and
300 (18) for a set of given parameters: T, σ_w, and ρ. According to Equations (12), R_{tb}
301 and R_{cb} also depend on the level of MH saturation, S_{MH}, which rules the average width
302 of bonds, B, hence the bond strength. The correlation between the S_{MH} and B will be

303 further explained in Section 3.3. Figure 10 illustrates how the strength envelopes
304 obtained for R_{sb0} and R_{rb0} vary with the level of MH saturation. The shape of the
305 envelopes which is controlled by the fitting parameters remains unchanged with the
306 variation of hydrate saturation, but the envelope size increases with the level of
307 hydrate saturation. The relationship between hydrate saturation and the average width
308 of bonds is further discussed in the next section.
309

310 **3.3 Geometric features of inter-particle MH**

311 From the experiments conducted by Hyodo et al. [32], it can be inferred that the
312 inter-particle MH bond dimension varies with the level of MH saturation. As shown
313 in Figure 2, a bond between cylindrical particles (or spherical ones in 3D) is
314 geometrically described by its width, B, and its thickness at the center, t_0. It is
315 convenient to define a dimensionless parameter β representing the ratio between
316 bond and particle sizes:

317 $$\beta = B/\overline{R} \tag{20}$$

318 In the literature, the hydrate saturation degree, S_{MH}, is defined as the ratio of the
319 methane hydrate volume to the total volume of the void. In the context of
320 two-dimensional problems such as in this study, S_{MH} reduces to the ratio of the
321 methane hydrate area A_{MH} (i.e., the area of void filled with bonds) to the total void
322 area A_V, i.e., $S_{MH} = A_{MH}/A_V$. The area of void occupied by the i-th bond (the blue area
323 as shown in Figure 2), A_{bi}, can be represented as a function of β from elementary
324 trigonometric considerations assuming that the radii of the two bonded particles are

equal to the equivalent radius, $\bar{R}$ (i.e. neglecting the different curvatures of the particles in the calculations):

$$A_{bi} = \bar{R}_i^2 \left[2\beta - \beta\sqrt{1-\frac{\beta^2}{4}} - 2\arcsin(\frac{\beta}{2}) + \frac{t_{0i}\beta}{\bar{R}_i} \right]. \quad (21)$$

The total area occupied by the bonds in a sample can be found by summation over all the bonds: $A_b = \sum_{i=1}^{m} A_{bi}$ with m being the total number of bonds, which depends on the initial configuration of the packing of the sample (i.e., whether loose or dense). Moreover, $A_{MH} = A_b + A_{MH_0}$ with A_{MH_0} being the area occupied by MH not binding any grains (i.e., floating around). Therefore, the level of hydrate saturation can be related to the area occupied by bonds as follows:

$$\begin{aligned} S_{MH} &= \frac{A_b + A_{MH0}}{A_V} = \frac{A_b}{A_V} + S_{MH_0} = \frac{(1+e_p)}{e_p A} A_b + S_{MH_0} \\ &= \frac{(1+e_p)}{e_p A} \sum_{i=1}^{m} \bar{R}_i^2 \left[2\beta - \beta\sqrt{1-\frac{\beta^2}{4}} - 2\arcsin(\frac{\beta}{2}) + \frac{t_{0i}\beta}{\bar{R}_i} \right] + S_{MH_0} \end{aligned} \quad (22)$$

where A is the total area of the sample; e_p is the planar void ratio; and S_{MH0} is the threshold value of hydrate saturation at which methane hydrates start to bond sand grains, which is around 20% - 30% [18, 19, 52, 53]. Note that the value of S_{MH0} depends on the hydrate growth history.

Equation (22) identifies a non-linear relationship that depends on the state of compaction of the sample (e.g., relative density), which rules the total coordination number of the sample and therefore the initial number of bonds, m. Figure 11 presents the relationship between $S_{MH} - S_{MH_0}$ and β achieved for the case of a dense sample (i.e., e_p= 0.21) at different critical values of bond thickness, t_0^{cr}, below which MH bonds form (i.e. bonds do not form for $t_0 > t_0^{cr}$). For an arbitrary value of t_0^{cr}, such

345 relationship can be used to determine the value of β to be employed in the
346 simulations for various levels of hydrate saturation S_{MH}.

347 Here, we want to replicate the natural process of formation of MH bonds as
348 closely as possible. The natural process of reference is given by MH bonds formed by
349 methane gas percolating through a deposit of sand particles. Hence, in our simulations,
350 once the DEM sample has been isotropically compressed to the confining pressure of
351 reference, bonds of various thickness were activated between particles with the
352 minimum bond thickness t_0 (see Figure 2) coinciding with the gap (the minimum
353 distance) between two adjacent particles.

354 It is important to note that MH bonds do not naturally exist in all the gaps of the
355 sample. In fact if the distance between two adjacent particles is larger than the
356 threshold value t_0^{cr}, there is little possibility for the formation of MH bonds. Figure
357 12(a) and (b) show typical SEM images of an artificial MHBS specimen with a MH
358 saturation of 50% and its host sand, respectively. We made an estimate of the
359 threshold distance t_0^{cr} based on several SEM images (as in Figure 12). After
360 identifying the outlines of the MHs and the particles of the host sand, we measured
361 the MH bond thickness t_0 at each particle contact. The measured value of t_0 was
362 always lower than 5% of d_{50}, with d_{50} being the median particle diameter. Hence, the
363 gap threshold for MH bond formation between two particles was taken as 5% of d_{50}.
364 It is worthy to mention that the value of t_0^{cr} may depend on factors such as MH
365 saturation, void ratio, type of host sand, which certainly need further investigation.
366 Here, for the sake of simplicity, we chose the same value of t_0^{cr} (i.e., 5% of d_{50}) in all

367 the simulations carried out.

368 Note that in the presented simulations once the bonds are broken they do not
369 reform. This implies that the time scale of the loading is much higher than the time
370 scale for bond formation so that MH induced bond formation during the occurrence of
371 the tests is negligible. For the investigation of phenomena such as MH extraction
372 induced submarine landslides and/or well instabilities, the assumption should be on
373 the conservative side since the reformation of MH bonds is expected to have a
374 stabilising effect opposing the development of failure mechanisms. However, in
375 principle it is possible to consider bond reformation in the analyses provided that an
376 evolution law for the bond strength over time, encapsulating the timescale of bond
377 reformation, is assigned. Equally, any other time dependent phenomena occurring at
378 the bond level could be considered by assigning a suitable time dependent bond law
379 (Nicot, 2004a, b).

380

381 # 4 DEM biaxial tests on MHBS

382 In this section, the 2D - DEM biaxial tests carried out to investigate the
383 macro-mechanical behaviors of MHBS are illustrated. Since in section 5 the DEM
384 results have been compared with the experimental results conducted by Masui et al.
385 [18], also a brief description of their test program is provided.

386

387 ## 4.1 DEM simulation procedure

388 The proposed bond contact model for MHBS was compiled by C++ code and

389 implemented into the commercial DEM software PFC2D [54] for the simulation of
390 biaxial compression tests. The simulations were carried out in three steps: (1)
391 generation of an un-bonded sample without cementation of MHs; (2) activation of
392 MH bonds in the sample; and (3) shearing of the sample under a pre-selected
393 confining pressure.

394 The 'multi-layer with under compaction method' proposed by Jiang et al. [55]
395 was used to generate a homogenous un-bonded sample consisting of 6000 particles at
396 a target void ratio of 0.21 which implies that the sample generated is dense. This
397 sample was then isotropically subjected to a confining pressure of 1 MPa, the exact
398 value applied in the tests conducted by Masui et al. [18]. Next, MH bonds were
399 activated in the sample at the confining pressure of 1 MPa at a given environmental
400 setting, i.e., surrounding temperature and backpressure. During the process of sample
401 generation, the wall-particle friction was set to zero to eliminate any boundary effects.
402 Figure 13 illustrates the DEM sample with a width of 400 mm and a height of 800
403 mm, in which the existence of the MH bonds is highlighted by black solid lines
404 linking adjacent particles. The amplified part of the sample in Figure 13 shows that
405 MH bonds have been successfully formed at all target contacts (i.e., whether virtual or
406 real contacts).

407 After sample preparation, the sample was sheared under a constant strain rate of
408 5% per minute by moving the top and bottom walls. The confining pressure was kept
409 constant at a pre-selected confining pressure during the shear phase by adjusting a
410 numerical servo-mechanism to the side walls.

411

412 **4.2 Sample parameters**

413 Figure 14 provides the grain size distribution of the DEM sample in comparison to
414 Toyoura sand adopted by Masui et al. [18]. The simulation used a more uniform
415 sample with particle diameter ranging from 6.0 mm to 9.0 mm in order to achieve
416 computational efficiency. The median diameter, d_{50}, of the DEM sample is 7.6 mm,
417 and the uniformity coefficient is 1.3 (i.e., the ratio of d_{60}/d_{10}, with d_{60} or d_{10} being the
418 particle diameter at which 60% or 10% of the mass of a soil sample is finer,
419 respectively).

420 Table 4 lists the parameters employed in the simulation. In case of un-bonded
421 particles, a trial-and-error procedure was used to determine the micro mechanical
422 parameters. Parameter were chosen in order to match as closely as possible the
423 material strength exhibited in triaxial tests on the host sand without any MHs.

424 In case of bonded particles, the parameters of the bond contact model (see Table
425 1) were determined via the relationships outlined in section 3 using as input the
426 experimental conditions (temperature, pressure, MH density. Etc.) of the experiments
427 run by Masui et al. [18]. The MH saturation degree used in the simulation was set to
428 be 0%, 25%, 40% and 55%. The pore-filling part of MH saturation (i.e., S_{MH0} in
429 Equation 22) was chosen as 20%. Accordingly, values of the bond area parameter β
430 at different MH saturations were obtained by Equation (22) (see Table 4). In Figure
431 11 the relationship between parameter β and MH saturation is provided. Note that
432 the relationship is specific to the PSD considered: if a different PSD is considered a

433 different relationship has to be expected. The surrounding temperature and water
434 pressure (i.e. backpressure applied to the sample) were $+5^0$C and 8.0 MPa,
435 respectively. Assuming a typical value of MH density, 0.9 g/cm^3, the Young's
436 modulus, the tension and compression strengths of MH were obtained by Equations
437 (9), (16) and (18), and listed in Table 4. Based on the parameters listed in Table 4, the
438 model parameters at each bond were computed according to Equations (6) and (12) by
439 a C++ subroutine. In Figure 10a and b the envelopes representing the strength of the
440 bond between two particles with average radius $\overline{R} = d_{50}/2$ subject to a tangential
441 force and a rolling moment respectively for MH saturations of 25%, 40% and 55% are
442 presented as an example. As shown in Figure 10, the value of all bond strengths (i.e.
443 R_{tb}, R_{cb}, R_{sb0}, R_{rb0}) increases as the MH saturation increases, showing an enhancement
444 of microscopic strength with MH saturation.
445

446 ## 5. DEM Simulation results

447 ### 5.1 Validation of the bond contact model

448 To check the validity of the bond contact model, numerically obtained
449 macro-mechanical behaviors (e.g., the stress-strain behavior and the volumetric
450 response) are presented here with respect to the MH saturation and effective confining
451 pressure in comparison to the experimental results obtained by Masui et al. [18].

452 Figure 15 presents the simulated stress-strain response of MHBS at four different
453 MH saturation degrees in comparison to the corresponding test results under the same
454 conditions. The comparison shows that the numerical simulations can effectively

455 replicate the influence of the MH saturation on stress-strain behaviors of MHBS for
456 the following aspects: (1) strain softening appears and becomes more and more
457 evident with the increasing of S_{MH}; (2) both the elastic modulus and maximum
458 deviator stress increase gradually with the increasing of S_{MH} and the axial strain at the
459 maximum deviator stress is around 2% ~ 3%, in good agreement with the
460 experimental results; (3) the variation of deviator stress with S_{MH} decays with the
461 axial stain after the deviator stress has reached the maximum value.

462 Based on the data in Figure 15, variations of the peak shear strength and the
463 elastic modulus of MHBS with respect to the MH saturation degree, S_{MH}, are provided
464 in Figure 16. Here, the peak shear strength is the deviator stress at its maximum value
465 (i.e., $\sigma_{peak} = (\sigma_1 - \sigma_3)_{\max}$), and elastic modulus, E_{50}, is the tangential modulus at the
466 point where the deviator stress is one half of the peak value. As shown in Figure 16,
467 the numerical results of the peak shear strength and elastic modulus increases
468 gradually with S_{MH}, which qualitatively agrees well with the experimental data.
469 Further, the variation of secant Young's modulus, E_{sec}, at various axial strains (e.g.,
470 0.5%, 1% and 1.5%) with MH saturation obtained from the numerical results is
471 presented in Figure 17. The secant modulus increases approximately linearly with S_{MH}
472 at all axial strains. Intercept of the lines increases with the axial strain while no
473 significant change is found in the slope. There is a large increase in E_{sec} at an axial
474 strain of 0.5%, when S_{MH} changes from 0% to 25%, indicating that the presence of
475 MH bonds greatly contributes to elastic modulus of the host sample, especially at
476 small axial strains. This is also denoted in Figure 15(b) that the initial slope of the

477 stress-strain relationship changes greatly when S_{MH} increases from 0% to 25%.

478 Figures 18(a) and (b) present the relationships between the volumetric strain and
479 the axial strain obtained from the experiments and numerical simulations, respectively.
480 It is clear that the volumetric strain in both numerical and experimental results exhibit
481 initial contractive behavior followed by shear dilation. Moreover the shear dilation
482 becomes evident with the increase of S_{MH} in both cases, showing a significant effect
483 of MH bonds on the volumetric dilative response. The variation of dilation angle with
484 S_{MH} is provided in Figure 19. The dilation angle in both experimental and numerical
485 cases gradually increases with S_{MH}. In addition, Figure 19(a) provides the dilation
486 angle obtained from numerical results at effective confining pressures of 2 MPa and 3
487 MPa in addition to 1 MPa. As shown in this figure, the dilation angle at the effective
488 confining pressure of 2 MPa and 3 MPa are also enhanced by S_{MH}, while this angle
489 decreases as the effective confining pressure increases.

490 Figure 20 provides an example of stress-strain behavior under different effective
491 confining pressures obtained by experiments and simulations at similar S_{MH}. The
492 stress-strain responses under these effective confining pressures exhibit
493 strain-softening with the maximum deviator stress being greatly enhanced by the
494 effective confining pressure. The variation of peak shear strength σ_{peak} with respect
495 to the effective confining pressure is provided in Figure 21, in which the relationships
496 with different S_{MH} are included. The peak shear strength increases dramatically as the
497 effective confining pressure increases at any S_{MH}. The data in Figure 21 lead to a
498 relationship between the peak strength parameters and S_{MH} as depicted in Figure 22.

In both numerical and experimental cases, the presence of MH causes considerable increase in cohesion, while no significant change is noted in the internal friction angle associated with the increasing of S_{MH}.

As discussed above, although the simulation cannot quantitatively reproduce the experimental tests, DEM implemented with the bond contact model can effectively capture the essential features relevant to the influence of the MH saturation degree and the effective confining pressure on the macro-mechanical behaviors of MHBS.

Constant volume (isochoric) tests were run for various values of the confining pressure and MH concentration to investigate the undrained behavior of the MHBS. As it is well known during the execution of an undrained test, excess pore pressures are generated so that a fully coupled liquid – solid numerical analysis would be necessary. However, (Shafipour and Soroush 2008) compared 2D fully coupled CFD – DEM analyses with 2D isochoric DEM analyses showing that constant volume DEM simulations are in good agreement so that it can be concluded that 2D isochoric DEM analyses may be used to investigate the material behavior in undrained conditions.

In figure xxx the results of undrained analyses run for various values of the confining pressure and MH concentration are shown. In all cases non liquefaction was exhibited. This is due to the fact that the samples considered were generated in loose conditions.

5.2 Microscopic information on bonds

521 Figure 23 provides the relationship between percentage of intact MH bonds and the
522 axial strain (0~30%) obtained from the DEM tests on the MHBS with different MH
523 saturation. Figure 23 shows that the percentage of intact bonds decreases with the
524 axial strain at a decreasing rate. The intact bonds diminish remarkably in particular
525 when the axial strain is lower 5%. This is the stage where the deviator stress drops
526 from the peak to a relatively low value as shown in Figure 15(b). Figure 24 also
527 presents the percentage of the intact bond area (i.e., A_{intact}/A_{total}, with A_{intact} being the
528 area of the intact bonds and A_{total} being the total area of the MH bonds). As shown in
529 Figure 23, the percentage of the intact bond area is a little higher than the percentage
530 of the intact bond number. This indicates that the MH bonds with relatively lower
531 area (i.e. the bonds with relatively lower width or between small particles) break more
532 likely than others during the shearing process. This is reasonable since bonds with a
533 lower width generally carry smaller bond strength.

534 Let us consider the sample with S_{MH} = 40% as an example to further clarify
535 microscopic information on bonds. Figure 24 presents the stress-strain and volumetric
536 strain response up to an axial strain of 30%, and the distribution of MH bonds at an
537 axial strain of 2%, 5%, 15%. In Figure 24(b), only intact MH bonds are marked as
538 black solid lines at particle contacts. Figure 24 shows that shear bands develop in the
539 sample and most of the bonds break in the bands. The percentage of broken bonds
540 increases with the expansion of shear bands when the axial strain grows. In addition,
541 when the axial strain reaches 15%, there are still some intact bonds left in the sample
542 (around 15% at S_{MH} = 40% as shown in Figure 23), and the intact bonded particles

543 outside the bands act as clumps. As a result, these intact bonds are expected to break
544 only if very large deformation of the sample occurs (e.g., 7% for S_{MH}=40% at axial
545 strain of 30% as shown in Figure 23). As indicated by Figure 24(a), the breakage of
546 remaining bonds contributes little to the deviator stress and volumetric strain of
547 MHBS when the axial strain reaches 15%. Hence, it can be concluded that the critical
548 state in terms of conventional definition can be reached while the bonds are not fully
549 broken. This may explain that the strength of the host sand coincides nearly with the
550 strength of the MH bearing sand in real experiments at an axial strain of around 15%
551 as shown in Figure 15.

552 Figure 25 presents the contact force chains for samples with MHs (S_{MH} = 40%)
553 and without MHs at the axial strain of 0% (onset of the shear phase), 2% (the peak
554 deviator stress) and 15% under the effective confining pressure of 1 MPa. The
555 thickness of the force chains is proportional to the value of contact forces, while the
556 direction represents the direction of the contact force. Figure 25 shows that the contact
557 force chains of the sample with MHs resemble to those without MHs at the onset of
558 the shear phase. This is because that the sample without activating the MH bonds had
559 reached the equilibrium under the confining pressure of 1 MPa. During the process of
560 bond activation, particles remained steady under such stress state and no significant
561 extra forces were resulted due to bond activation. However, the stress state of the
562 samples changes gradually due to the vertical loading during the shear process. The
563 contact force chains exhibit differently in two cases, because MH bonds can bear
564 additional inter-particle forces caused by loading than contacts without bonds. As

565 shown in Figure 25 (a) and (b), tension forces marked as the red lines appear in the
566 samples with MHs, while only compression force are found at the particle contacts in
567 the samples without MHs. Besides, the compression force of the samples with MHs is
568 larger than that without MHs due to the contribution of MH bonds. When the axial
569 strain increases to 15%, the contact force chains of sample with MH bonds resemble
570 to those without MH bonds. This further indicates that the strength of the MHBS
571 tends to be close to the strength of the host sand associated with the increasing of
572 axial strain due to the large number of bond breakage (see Figure 24).
573

574 **5.3 Discussion**

575 The DEM simulation incorporating the bond contact model of MHBS can capture
576 several key features of MHBS, particularly the bonding effects of MHs, providing an
577 effective tool to understand the micro-mechanism of the mechanical behaviors of
578 MHBS. However, some differences were identified between the numerical and the
579 experimental data [18].

580 A 2D model was used in the present study instead of 3D models due to several
581 reasons. First of all there is no available experimentally-validated contact law for
582 three-dimensional cemented granules. Many efforts were made in the past several
583 years to develop reliable equipment and repeatable procedure for conducting a micro
584 mechanical experiment to validate the bond contact law [43, 44, 56]. However this
585 work was only restrained to 2D context using rods, because the test on spheres in
586 three dimensions will become extremely difficult in particular when a complex of

loads is of desire. Second, DEM simulations of engineering problems of interest, e.g., landslides along the seabed or well instabilities induced by MH extraction, are computationally very expensive and hence are only possible in 2D for the foreseeable future. Third, though 3D modeling can provide more realistic results, the 2D model can be still used as a useful research tool due to simplicity in implementation and easy interpretation of results. Moreover, 2D simulation can still shed light on the importance of MH bonds in MHBS and the growing pattern of shear bands in MHBS, which will not significantly differ from that in 3D modeling. Although 2D modeling has limit to quantitatively replicate volumetric responses obtained from the 3D triaxial tests, it can still produce realistic strength parameters, particularly the friction angle, which has been demonstrated in Figure 22.

Granules of the host sand were idealized as disc-shaped particles, which differ from the realistic soil gains with irregular shapes. Excessive rotation will be resulted from circular particles if no particular action is taken to improve the contact model. In order to imitate the realistic shapes of soil grains and, more importantly, produce realistic bulk strength of soils, a few techniques are now available, such as clustered discs [57], polygon [58], polyarc elements [59] among others (e.g., [60]), all of which can reduce excess particle rotation. However, using particles with irregular shapes will give rise to some practical issues in addition to the loss in computational efficiency due to demand for a rigorous contact detection algorithm and others. For instance, extreme effort will be certainly needed to accurately generate even a small sample with exact microstructure (e.g., shape and fabric) as real soils. Moreover,

609 particle angularity will evolve during loading process, and such morphological change
610 becomes almost untraceable in a practical simulation using particles of sufficient
611 number. Alternatively, realistic particle rotation can still be achieved from
612 disc-shaped particles by directly incorporating the inter-particle rolling resistance into
613 the contact law in the manner that this present study adopted (e.g., [45, 61, 62]). This
614 approach sacrifices details at the particle scale such as the particle shape, but is
615 particularly suitable where accurate bulk behaviors of the medium are prior to fidelity
616 to the actual particle shapes.

617 The grain size distribution of the DEM sample differs from that of Toyoura sand
618 used in the experiment of Masui et al. [18]. The average particle diameter of the DEM
619 sample (i.e., 7.6 mm) is larger than that of Toyoura sand (i.e., below 0.2 mm). Larger
620 particles were used in this study for computational efficiency. As long as the model
621 parameters are well calibrated following an appropriate procedure, an assemblage of
622 large particles can still replicate realistic mechanical properties of soils, which is of
623 the most value among other particle characteristics for the goal of this paper. This has
624 been demonstrated in Figures 15 to 22. Note that this bond contact model and the
625 procedure for determining the model parameters presented here are also applicable to
626 MHBS samples with finer particles except that the best-fit parameter set may be
627 different from the one presented herein due to the differences in the particle size.

628 This work has a few limitations. The focus of this study was only on the MHs
629 acting as the cementation agent at inter-particle contacts in MHBS. The contribution
630 from other types of MHs, such as pore-filling and coating, may be under- or

631 over-estimated. An arbitrary value was assigned to the threshold value of the hydrate
632 saturation degree, below which only pore-filling MHs will form and this type of MHs
633 is assumed to carry negligible effects on the bulk mechanical properties of MHBS.
634 However, pore-filling MHs may have some effect on the strength of MHBS since they
635 can bear and transmit loads as the soil skeleton deforms. This kind of effect was
636 revealed by Brugada et al.[8]. It is noted that simultaneous consideration of different
637 types of MHs will become meaningful in DEM analyses only if identification and
638 quantification of different types of MHs become possible in experiment with the
639 advancement of microstructure detection techniques.
640

641 ## 6 Conclusions

642 This study proposed a two-dimensional bond contact model for the DEM simulation
643 of soils with methane hydrates (MHs) cementing at inter-particle contacts of grains. A
644 conceptual bond contact model [36], which has been validated through a
645 specifically-designed test [43, 44], was first introduced with an emphasis on the
646 generic contact law, which was later adapted in MHBS. Due to difficulty in
647 conducting the micro-mechanical tests on bonded granules using real MHs, the model
648 parameters in relation to the bond strength and particle stiffness were suggested to be
649 formulated from the correlation to the surrounding temperature, water pressure and
650 MH density. The bond width and thickness varying at each bond were related to a
651 pre-selected value of MH saturation degree. With the aid of a customized C++
652 subroutine, the proposed model was successfully implemented into a DEM software,

PFC2D, and then testified through a series of biaxial compression tests on MHBS at various levels of MH saturation and different surrounding environmental conditions (i.e., different temperatures and pressures).

Compared with the experimental data obtained by Masui et al. [18], the DEM simulation can effectively capture the major mechanical behaviors of MHBS, e.g., the phenomenon of strain softening and shear dialation which become evident with the increasing of the hydrate saturation degree. In particular, the model is able to reproduce realistic strength properties of MHBS samples at a wide range of the hydrate saturation degree, which is in good agreement with the experimental results. Although some quantitative differences with the experimental triaxial tests were observed, the proposed bond model and its parameter calibration proved to be realistic so that the proposed bond model can be employed for DEM simulations of problems of engineering interests.

The proposed model can also serve as a useful tool to better understand the connection between the microscopic behaviors and the macroscopic properties of MHBS samples. The presented set of simulations shed light on the mechanical behavior of the MH bearing sands at the microscopic level, e.g., spatial distribution of bond breakage, type of bond failure (either shear or tensile), redistribution of forces within contact chains so that our understanding is clearer on the relationship between cementation and de-bonding mechanisms occurring at the micro level and macro-mechanical behavior of MHBS.

Acknowledgments

This research was financially supported by China National Funds for Distinguished Young Scientists (Grant No. 51025932), National Natural Science Foundation of China (Grant No. 51239010), and Ph.D Programs Foundation of Ministry of Education of China with (Grant No. 20100072110048). These joint supports are sincerely appreciated.

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